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VOID FORMATION IN HEAVILY IRRADIATED NaCl

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Experimental and theoretical results are presented on the formation of voids discovered recently in heavily irradiated NaCl. Experimental data show that for NaCl samples with particular dopants, the development of radiation damage, such as Na-colloids, chlorine inclusions and voids does not show a saturation as a function of the dose. A quantitative comparison of the new model (Dubinko *et al.*, These proceedings) for radiation damage in alkali halides with experimental data is presented. Mean sizes and volume fractions of all types of observed defects are calculated. It is shown that voids formed due to agglomeration of F centers and cation vacancies can grow to the dimensions exceeding the mean distance between colloids and bubbles, eventually absorbing them, and, hence, bringing the halogen gas and metal to a back reaction. Impurities are shown to play a major role in the void development with increasing irradiation dose, which strongly affects the radiation stability of NaCl.

Keywords: Radiation damage; NaCl; Vacancy voids

1. INTRODUCTION

The natural and doped NaCl crystals have been irradiated with 1.35 MeV electrons up to fluences 6×10^{18} electrons/cm^2 that corresponded to doses of 150 Grad (about 30 displacements per atom) at

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temperatures between 50°C and 150°C. Scanning electron microscopy (SEM) in combination with differential scanning calorimetry (DSC) and Electron Spin Resonance (ESR) were used to study the void production. The concentration of the metallic Na was deduced from measurements of the latent heat of melting of metallic Na in combination with ESR.

Formation of large voids (above 100 nm in diameter) correlates with an eventual destruction of the samples under heavy irradiation or heating without any observable loss of the specimen weight, which confirms that the cavities are vacancy voids rather than gas-filled bubbles. As can be seen from Fig. 1, impurities strongly affect the void development as well as the accumulation of metallic Na.

A quantitative comparison of a new model of radiation damage in alkali halides, described in the companion paper [1] with experimental data, is presented below.

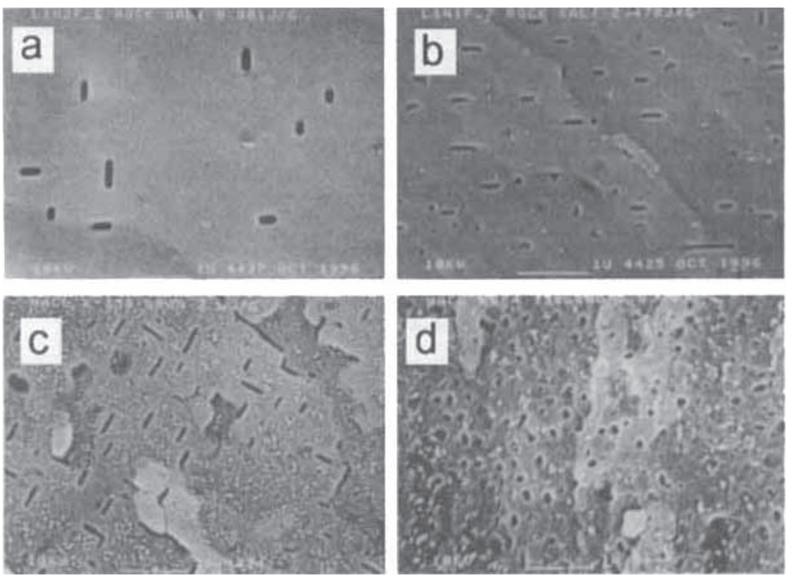

FIGURE 1 SEM micrographs showing vacancy voids in NaCl samples irradiated with electrons to a dose of 150 Grad. (a) and (b) natural rock salt samples irradiated at 140°C and 70°C, LHM of metallic Na is 0.8 J/g and 2.5 J/g, respectively; (c) NaCl + K (0.1 mol%) irradiated at 100°C, LHM of metallic Na is 3.6 J/g; (d) NaCl + KBF_4 (0.03 mol%) irradiated at 100C, LHM of metallic Na is 5.6 J/g.

2. COMPARISON OF THE MODEL PREDICTIONS WITH EXPERIMENTAL DATA

Figure 2 illustrates the radiation-induced reactions between point defects (PD) and extended defects (ED) based on the present model [1]. Primary radiation PD, namely, H and F centers, separate ultimately into bubbles, dislocations and metal colloids, which results in production of the secondary PD (cation vacancies) and ED (interstitial loops and vacancy voids). Based on the model, a complete set of the rate equations for PD and growth rates for ED was derived, which is described in detail elsewhere [2]. An asymptotic (in time of irradiation) solution to these equations was obtained [2] in the temperature range above $0.3T_m$ (T_m is the melting point), in which both anion and cation PD are mobile, but below the temperature T^{th}, at which thermal

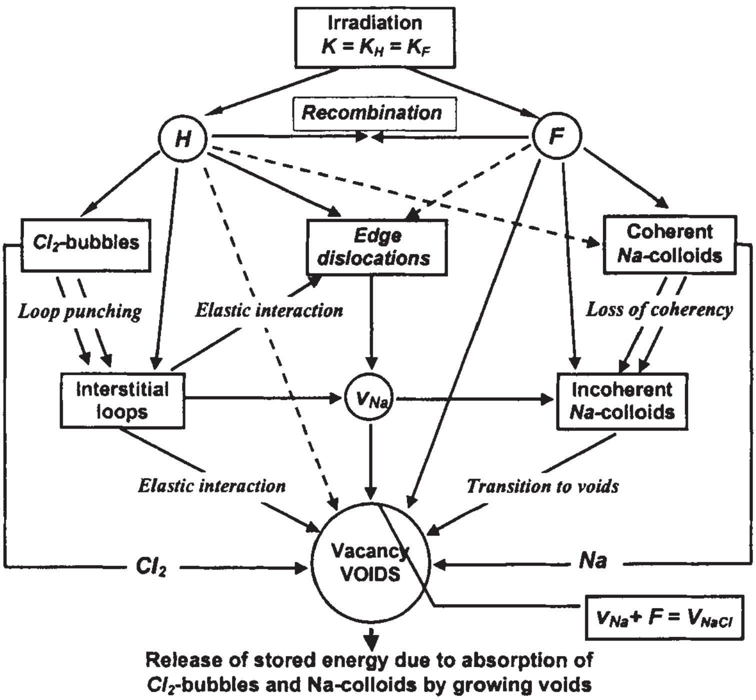

FIGURE 2 Diagram of radiation-induced reactions between point defects (H- and F-centers, and cation vacancies) and extended defects (bubbles, dislocations and colloids) resulting in the void formation.

TABLE I Principal material and irradiation parameters used for the calculations in the present paper

Parameter	*Value*	*Parameter*	*Value*
Dislocation bias, δ_d	0.5	Irradiation temperature, T, °C	100
$\delta_{eff} = \delta_d - \alpha_\mu(\sigma_\varepsilon/\mu)^2$	0.1	Dose rate, K, Mrad/h (dpa/s)	120 (6×10^{-6})
"Image" interaction constant, α_{im}	0.1	RIC upper temperature, T^{th} (K), °C	220
Modulus interaction constant, α_μ	100	Maximum dose, Grad (dpa)	150 (30)
Shear modulus, μ, GPa	15	F–H recombination constant, m^{-2}	10^{20}
Colloid shear modulus, μ_C, GPa	3.3	Dislocation density, ρ, m^{-2}	1×10^{14}–5×10^{14}
Colloid misfit, ε	0.05	Void nucleation rate, J_v, $dpa^{-1}\,m^{-3}$	10^{18}–10^{19}

evaporation of PD from ED starts to play essential role and suppress the growth of ED. In this region, the growth or shrinkage rates of different kinds of ED and their sizes are determined by the difference between the incoming fluxes of radiation produced PD, which is determined by several material constants presented in Table I. We have used experimentally observed values for the mean dislocation density, ρ, and the void nucleation rate, J_v, as the only input microstructural parameters (see Table I). The mean sizes and number densities of colloids and bubbles as well as the mean size of voids have been calculated as a function of irradiation dose at the stage when the nucleation stage of colloids is over. Under this condition, the colloid number density is independent of the initial conditions and is determined by the radiation-induced coarsening (RIC) mechanism, as was originally proposed for voids in irradiated metals [3].

Figure 3(a) shows the measured dose dependence of the latent heat of melting (LHM) of Na (which is proportional to the total content of metallic Na) in samples doped with different impurities along with the theoretical curves corresponding to different dislocation densities. Figure 3(b)–(d) shows the measured and calculated dependencies of void parameters on LHM. The mean sizes and number densities of colloids and bubbles were not measured, while their maximum theoretical values were found to be about 6 nm and $10^{23}\,m^{-3}$, respectively, as compared to 60 nm and $5 \times 10^{19}\,m^{-3}$ for voids.† For doses higher than

† Note that in the Fig. 3(c), the void number density was measured at the sample surface, and the value $5 \times 10^{19}\,m^{-3}$ is the corresponding void number per unit volume.

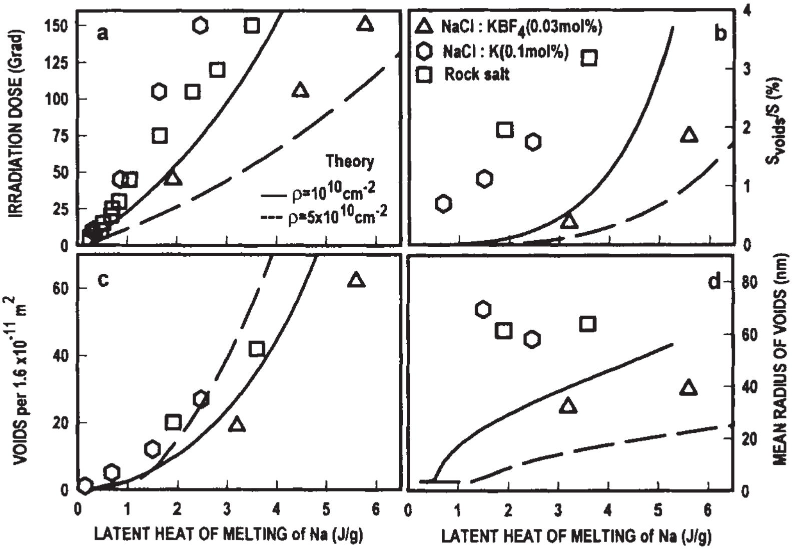

FIGURE 3 Measured and calculated dependence of LHM of metallic Na on irradiation dose (a), and the void mean parameters against LHM for different dopants and dislocation densities, ρ. Symbols – experimental data, curves – theory.

50 Grad, the void dimensions exceed the mean distance between colloids and bubbles, which makes possible the capture of the latter by growing voids that would eventually bring the halogen gas and metal to a back reaction, as discussed in the previous paper [1].

Acknowledgment

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